

## Supplementary Materials

### Large tunneling magnetoresistance in room-temperature magnetic tunnel junctions based on altermagnetic $XV_2Y_2O$

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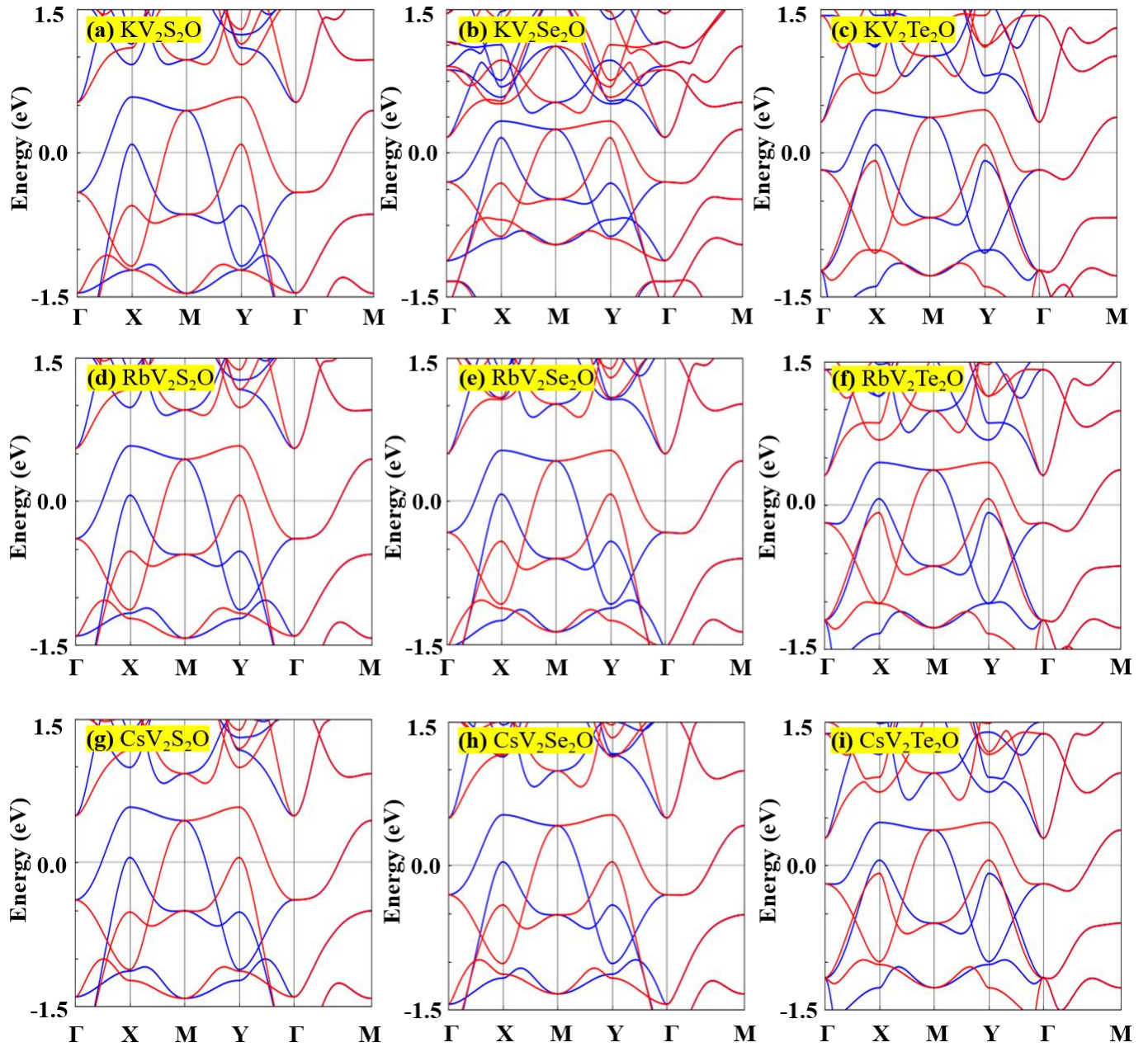
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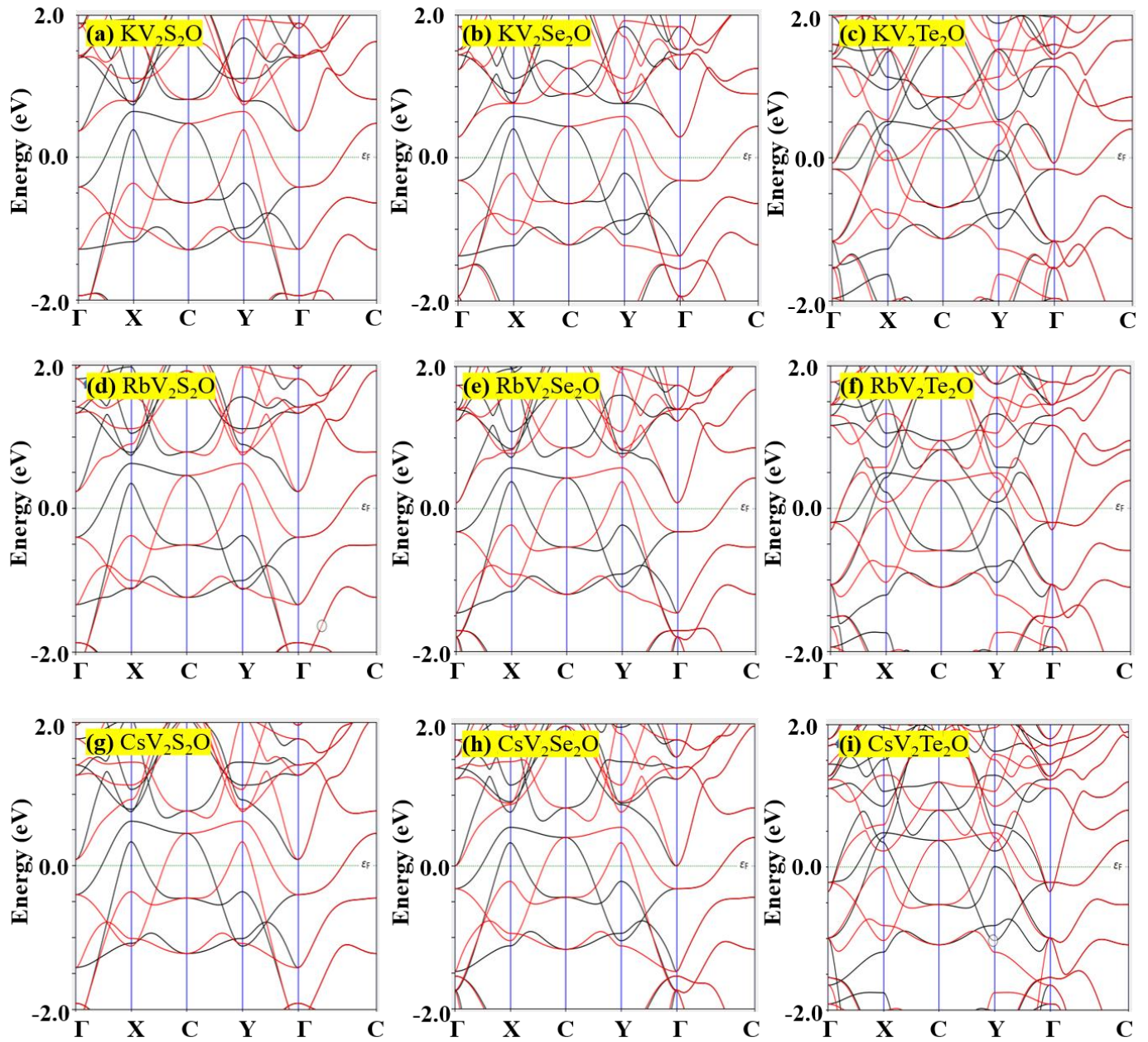
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|                                                                                                                                                                                                                                                                                                                                                       |                                                                                                                                                                                                                                                                                                                                                        |                                                                                                                                                                                                                                                                                                                                                              |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <b>KV2S2O (a)</b><br>1.000000<br>3.900606 0.000000 0.000000<br>0.000000 3.900606 0.000000<br>0.000000 0.000000 6.982870<br>K V S O<br>1 2 2 1<br>Direct<br>0.000000 -0.000000 0.500000<br>0.500000 -0.000000 0.000000<br>0.000000 0.500000 0.000000<br>0.500000 0.500000 0.230083<br>0.500000 0.500000 0.769917<br>0.000000 -0.000000 -0.000000       | <b>KV2Se2O (b)</b><br>1.000000<br>3.951940 0.000000 0.000000<br>0.000000 3.951940 0.000000<br>0.000000 0.000000 7.352430<br>K V Se O<br>1 2 2 1<br>Direct<br>0.000000 0.000000 0.500000<br>0.500000 0.000000 0.000000<br>0.000000 0.500000 0.000000<br>0.500000 0.500000 0.240450<br>0.500000 0.500000 0.759550<br>0.000000 0.000000 0.000000          | <b>KV2Te2O (c)</b><br>1.000000<br>4.027171 0.000000 0.000000<br>0.000000 4.027171 0.000000<br>0.000000 0.000000 8.227060<br>K V Te O<br>1 2 2 1<br>Direct<br>0.000000 0.000000 0.500000<br>0.500000 0.000000 0.000000<br>0.000000 0.500000 0.000000<br>0.500000 0.500000 0.239183<br>0.500000 0.500000 0.760817<br>0.000000 0.000000 0.000000                |
| <b>RbV2S2O (d)</b><br>1.000000<br>3.930690 0.000000 0.000000<br>0.000000 3.930690 0.000000<br>0.000000 0.000000 7.171010<br>Rb V S O<br>1 2 2 1<br>Direct<br>-0.000000 -0.000000 0.500000<br>0.500000 -0.000000 -0.000000<br>-0.000000 0.500000 -0.000000<br>0.500000 0.500000 0.222198<br>0.500000 0.500000 0.777802<br>-0.000000 -0.000000 0.000000 | <b>RbV2Se2O (e)</b><br>1.000000<br>3.965218 0.000000 0.000000<br>0.000000 3.965218 0.000000<br>0.000000 0.000000 7.759473<br>Rb V Se O<br>1 2 2 1<br>Direct<br>0.000000 0.000000 0.500000<br>0.500000 0.000000 -0.000000<br>0.000000 0.500000 -0.000000<br>0.500000 0.500000 0.225406<br>0.500000 0.500000 0.774594<br>0.000000 0.000000 0.000000      | <b>RbV2Te2O (f)</b><br>1.000000<br>4.043609 0.000000 0.000000<br>0.000000 4.043609 0.000000<br>0.000000 0.000000 8.449990<br>Rb V Te O<br>1 2 2 1<br>Direct<br>0.000000 -0.000000 0.500000<br>0.500000 -0.000000 0.000000<br>0.000000 0.500000 0.000000<br>0.500000 0.500000 0.231251<br>0.500000 0.500000 0.768749<br>0.000000 -0.000000 0.000000           |
| <b>CsV2S2O (g)</b><br>1.000000<br>3.940954 0.000000 0.000000<br>0.000000 3.940954 0.000000<br>0.000000 0.000000 7.488521<br>Cs V S O<br>1 2 2 1<br>Direct<br>0.000000 0.000000 0.500000<br>0.500000 0.000000 -0.000000<br>0.000000 0.500000 -0.000000<br>0.500000 0.500000 0.211635<br>0.500000 0.500000 0.788365<br>0.000000 0.000000 0.000000       | <b>CsV2Se2O (h)</b><br>1.000000<br>4.007704 0.000000 0.000000<br>0.000000 4.007704 0.000000<br>0.000000 0.000000 7.875740<br>Cs V Se O<br>1 2 2 1<br>Direct<br>-0.000000 -0.000000 0.500000<br>0.500000 -0.000000 0.000000<br>-0.000000 0.500000 0.000000<br>0.500000 0.500000 0.218340<br>0.500000 0.500000 0.781660<br>-0.000000 -0.000000 -0.000000 | <b>CsV2Te2O (i)</b><br>1.0000000000000000<br>4.057980 0.000000 0.000000<br>0.000000 4.057980 0.000000<br>0.000000 0.000000 8.786550<br>Cs V Te O<br>1 2 2 1<br>Direct<br>-0.000000 0.000000 0.500000<br>0.500000 0.000000 0.000000<br>-0.000000 0.500000 0.000000<br>0.500000 0.500000 0.221230<br>0.500000 0.500000 0.778770<br>-0.000000 0.000000 0.000000 |

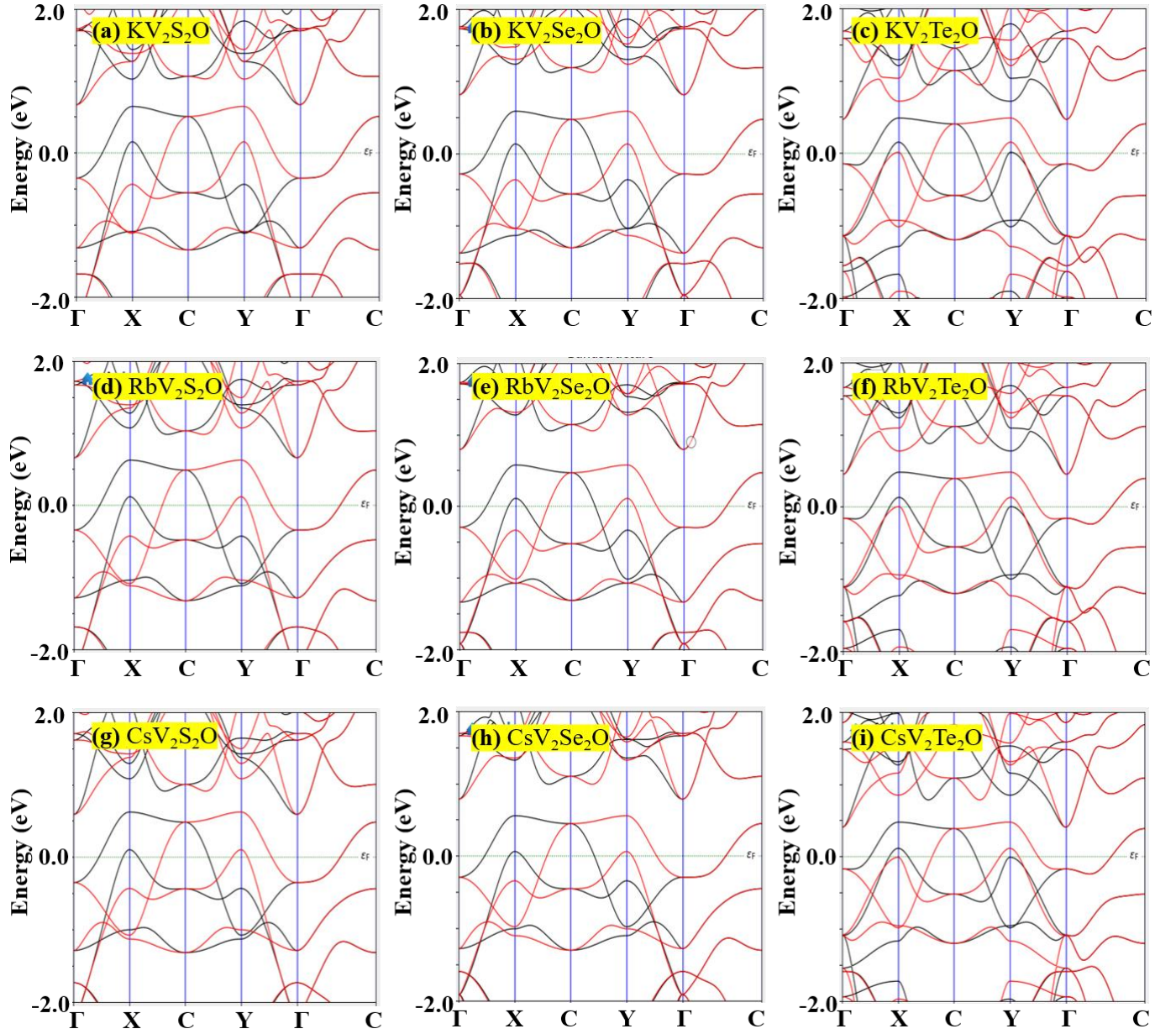
**Fig. S1. The optimized POSCAR configurations of altermagnetic  $XV_2Y_2O$  materials.** (a-i) The relaxed lattice constant of altermagnetic  $XV_2Y_2O$  family ( $X = K, Rb, Cs$ ;  $Y = S, Se, Te$ ).



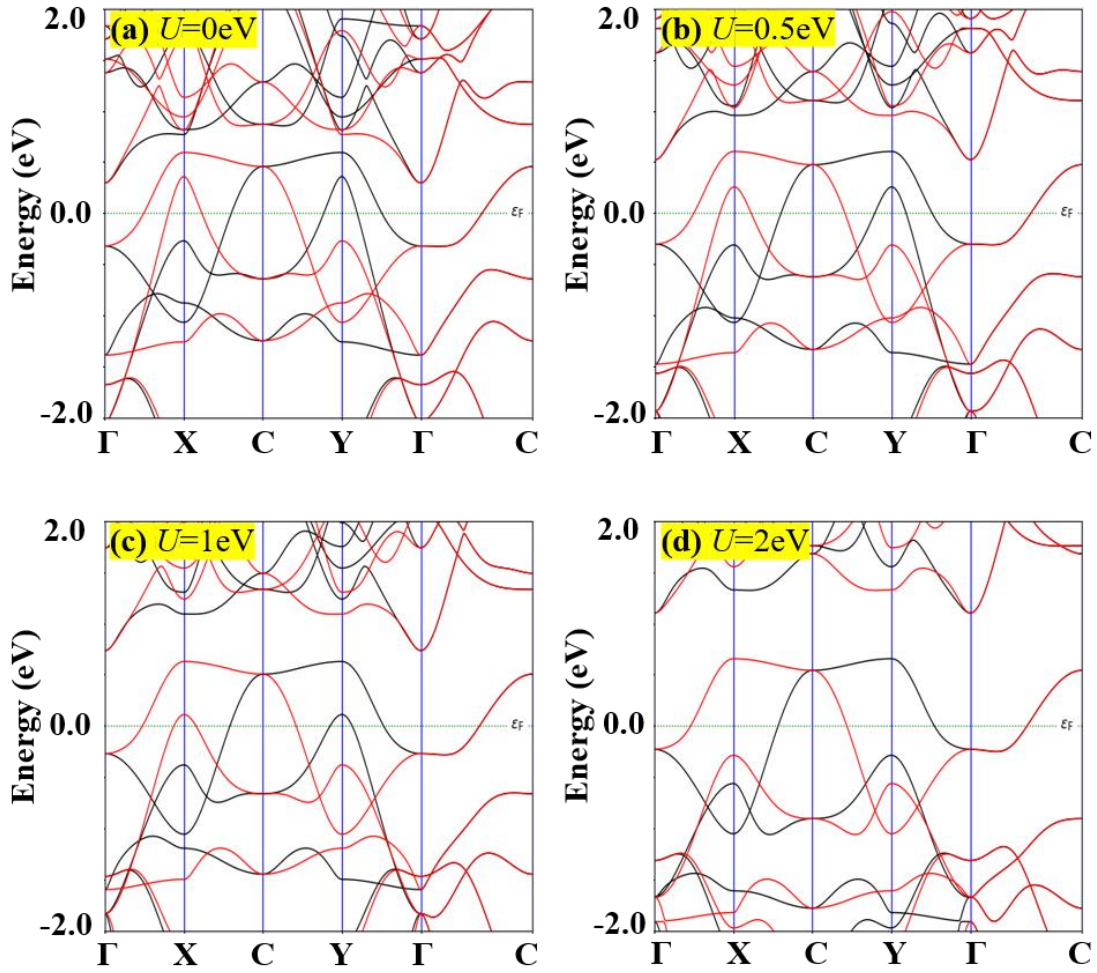
**Fig. S2.** Band structures of altermagnetic  $XV_2Y_2O$  materials calculated using VASP. (a-i) Spin-resolved band for  $XV_2Y_2O$  ( $X=K, Rb, Cs$ ;  $Y=S, Se, Te$ ) calculated using VASP with GGA+ $U$  ( $U = 1$  eV). Red and blue lines denote spin-up and spin-down channels, respectively.



**Fig. S3. Band structures of altermagnetic  $XV_2Y_2O$  materials obtained with FHI pseudopotentials.** (a-i) Spin-resolved band for  $XV_2Y_2O$  ( $X=K, Rb, Cs$ ;  $Y = S, Se, Te$ ) calculated using QuantumATK with FHI pseudopotentials and a single-zeta-polarized basis set.



**Fig. S4. Band structures of altermagnetic  $XV_2Y_2O$  materials obtained with PseudoDojo pseudopotentials.** (a-i) Spin-resolved band for  $XV_2Y_2O$  ( $X=K, Rb, Cs$ ;  $Y = S, Se, Te$ ) calculated using QuantumATK with PseudoDojo pseudopotentials, a “Medium” basis set, and SGGA+ $U$  ( $U = 0.5$  eV)



**Fig. S5. Band structures of  $\text{KV}_2\text{Se}_2\text{O}$  on the Hubbard  $U$  parameter.** (a-d) Spin-resolved band for  $\text{KV}_2\text{Se}_2\text{O}$  calculated using QuantumATK with FHI pseudopotentials, a single-zeta-polarized basis set, and SGGA+ $U$  ( $U=0, 0.5$  eV,  $1$  eV,  $2$  eV).

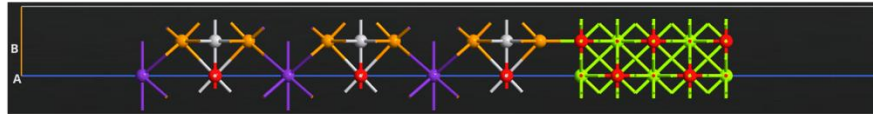
**Tab. S1. The Transmission spectrum and TMR value of  $XV_2Y_2O/n$ -Layers MgO/ $XV_2Y_2O$  MTJ devices.**

| Device type                                     | Layers      | $T_P$     | $T_{AP}$  | TMR (%) |
|-------------------------------------------------|-------------|-----------|-----------|---------|
| <b>RbV<sub>2</sub>Te<sub>2</sub>O-based MTJ</b> | 3-Layer MgO | 6.404e-07 | 1.282e-10 | 499411  |
|                                                 | 4-Layer MgO | 5.888e-09 | 1.423e-11 | 41278   |
|                                                 | 5-Layer MgO | 4.868e-09 | 2.810e-12 | 173134  |
|                                                 | 6-Layer MgO | 1.894e-09 | 1.840e-13 | 1029229 |
|                                                 | 7-Layer MgO | 1.026e-09 | 3.162e-14 | 3246321 |
| <b>KV<sub>2</sub>Se<sub>2</sub>O-based MTJ</b>  | 3-Layer MgO | 2.496e-06 | 1.104e-09 | 225893  |
|                                                 | 4-Layer MgO | 7.966e-08 | 6.611e-10 | 11948   |
|                                                 | 5-Layer MgO | 6.611e-08 | 3.422e-10 | 19217   |
|                                                 | 6-Layer MgO | 1.981e-08 | 2.126e-11 | 93089   |
|                                                 | 7-Layer MgO | 3.935e-09 | 5.460e-14 | 7206907 |

**Tab. S2. The lattice mismatch between  $XV_2Y_2O$ , the  $X_2Y$  barrier and the metal Ag. The sections marked in red indicate the strained layer.**

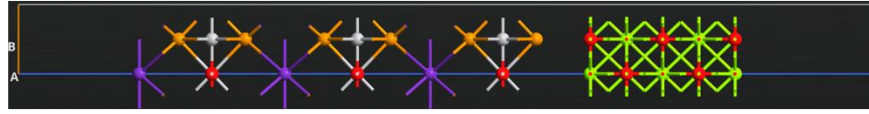
|                                                       | Mismatch |                                       | Mismatch |
|-------------------------------------------------------|----------|---------------------------------------|----------|
| KV <sub>2</sub> S <sub>2</sub> O-K <sub>2</sub> S     | 3.5%     | Ag-KV <sub>2</sub> S <sub>2</sub> O   | 3%       |
| KV <sub>2</sub> Se <sub>2</sub> O-K <sub>2</sub> Se   | 2%       | Ag-KV <sub>2</sub> Se <sub>2</sub> O  | 2.2%     |
| KV <sub>2</sub> Te <sub>2</sub> O-K <sub>2</sub> Te   | 1%       | Ag-KV <sub>2</sub> Te <sub>2</sub> O  | 0.96%    |
| RbV <sub>2</sub> S <sub>2</sub> O-Rb <sub>2</sub> S   | 0.72%    | Ag-RbV <sub>2</sub> S <sub>2</sub> O  | 2.5%     |
| RbV <sub>2</sub> Se <sub>2</sub> O-Rb <sub>2</sub> Se | 1.1%     | Ag-RbV <sub>2</sub> Se <sub>2</sub> O | 2%       |
| RbV <sub>2</sub> Te <sub>2</sub> O-Rb <sub>2</sub> Te | 0.78%    | Ag-RbV <sub>2</sub> Te <sub>2</sub> O | 1.1%     |
| CsV <sub>2</sub> S <sub>2</sub> O-Cs <sub>2</sub> S   | 2.9%     | Ag-CsV <sub>2</sub> S <sub>2</sub> O  | 2.4%     |
| CsV <sub>2</sub> Se <sub>2</sub> O-Cs <sub>2</sub> Se | 3.8%     | Ag-CsV <sub>2</sub> Se <sub>2</sub> O | 1.3%     |
| CsV <sub>2</sub> Te <sub>2</sub> O-Cs <sub>2</sub> Te | 6.4%     | Ag-CsV <sub>2</sub> Te <sub>2</sub> O | 0.45%    |

(a)  $\text{RbV}_2\text{Te}_2\text{O}/\text{MgO}$



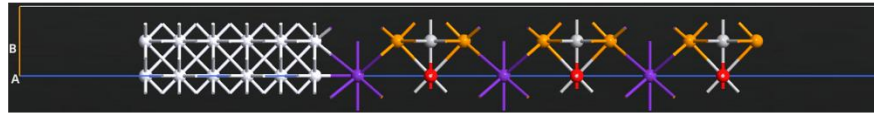
$$32.5514 \text{ Å} - 30.1419 \text{ Å} = 2.4095 \text{ Å}$$

**Relaxed:**



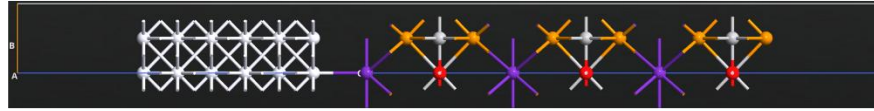
$$33.3123 \text{ Å} - 30.1419 \text{ Å} = 3.1704 \text{ Å}$$

(b) Metal/ $\text{RbV}_2\text{Te}_2\text{O}$



$$19.5871 \text{ Å} - 17.1062 \text{ Å} = 2.4809 \text{ Å}$$

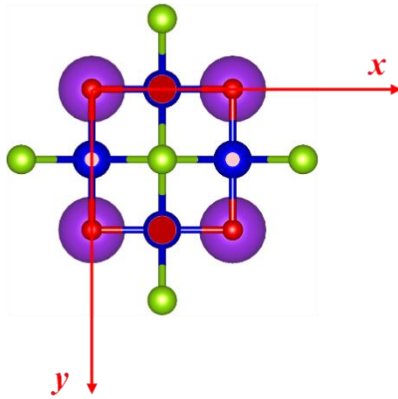
**Relaxed:**



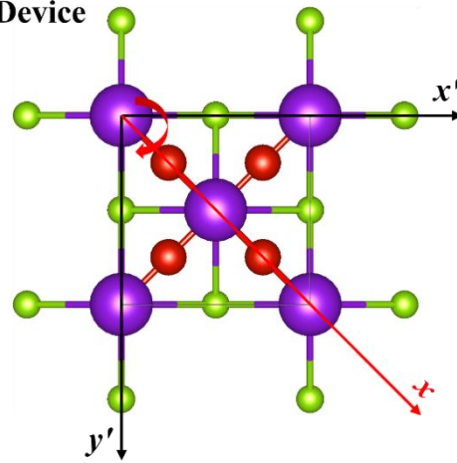
$$20.1909 \text{ Å} - 17.1062 \text{ Å} = 3.0847 \text{ Å}$$

**Fig. S6. The relaxed device structures of  $\text{RbV}_2\text{Te}_2\text{O}$  heterostructures.** Initial and relaxed atomic geometries of (a) the  $\text{RbV}_2\text{Te}_2\text{O}/\text{MgO}$  and (b) the Metal/ $\text{RbV}_2\text{Te}_2\text{O}$  contact configuration.

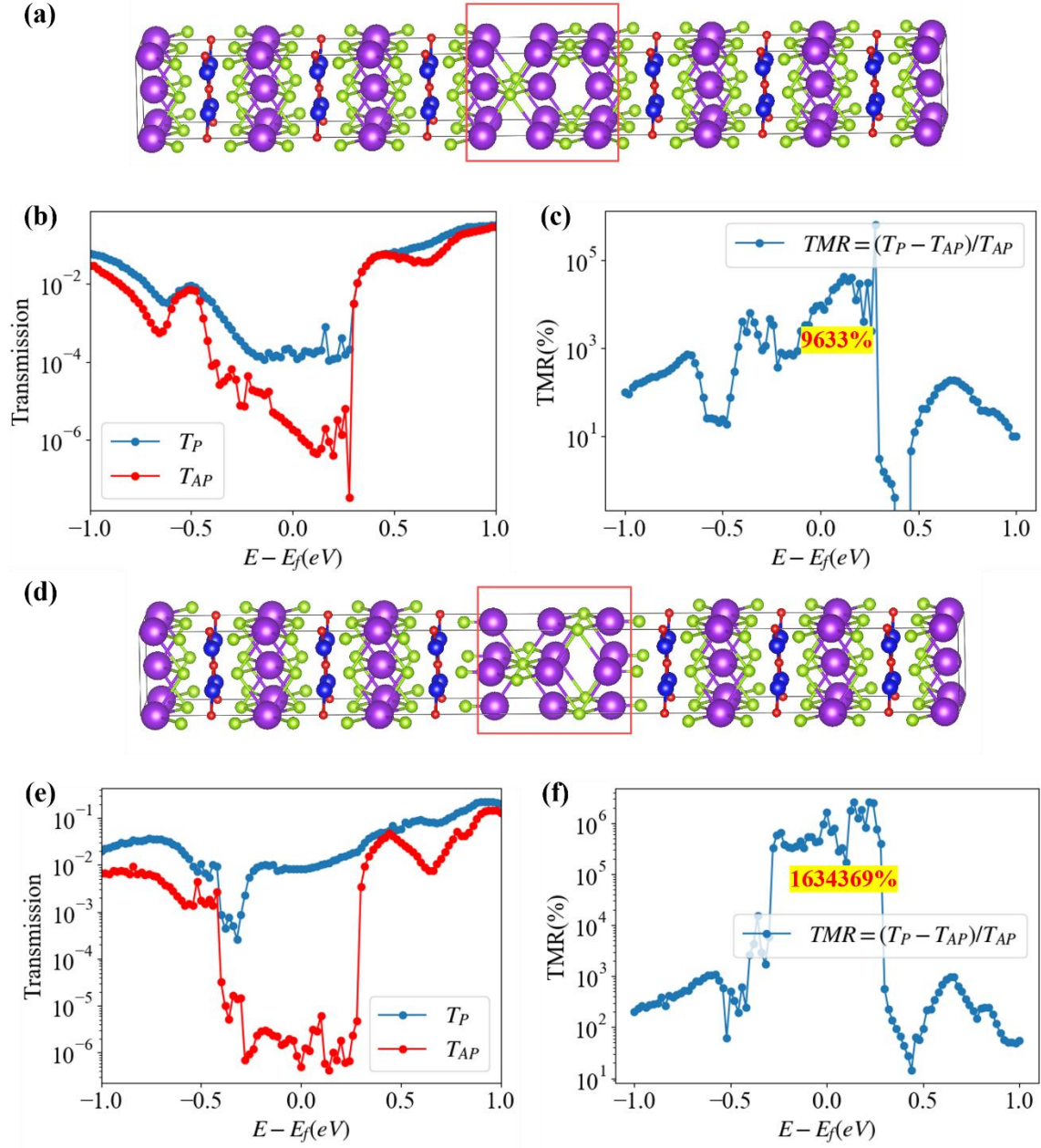
(a) Bulk



(b) Device



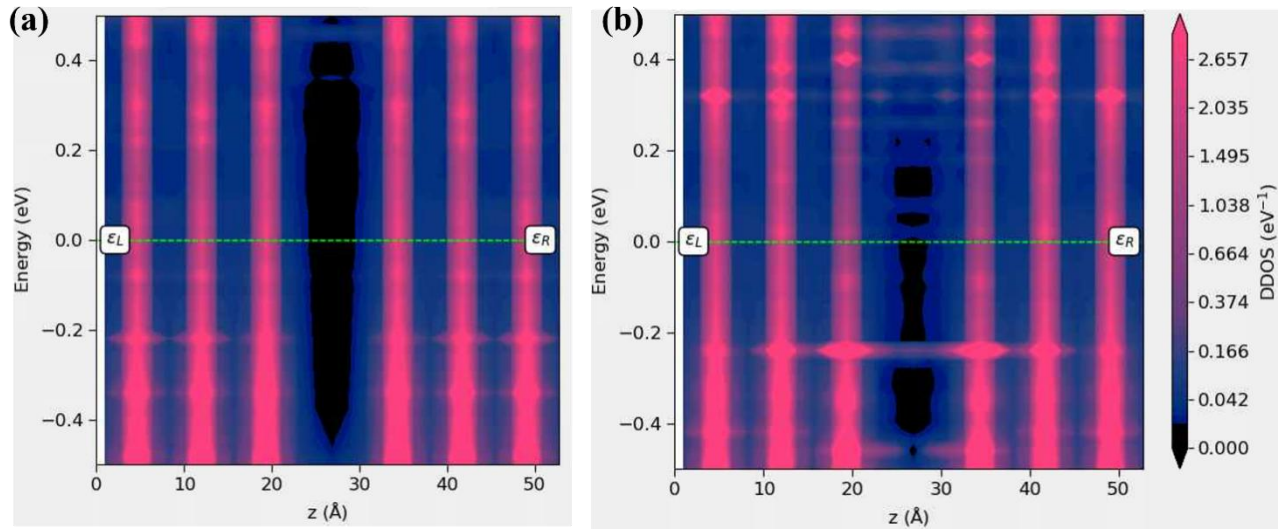
**Fig. S7. Crystal orientation alignment and supercell construction for devices.** (a) The top view of atomistic bulk  $\text{XV}_2\text{Y}_2\text{O}$  and  $\text{XV}_2\text{Y}_2\text{O}$ -based MTJ devices; (b) Atomic setup of  $\text{XV}_2\text{Y}_2\text{O}$ -based MTJ device supercell, where a  $45^\circ$  rotation to the coordinate system relative to the crystallographic axes of the bulk  $\text{XV}_2\text{Y}_2\text{O}$  was applied to ensure lattice matching.



**Fig. S8. Device structures and spin transport properties of KV<sub>2</sub>Se<sub>2</sub>O/K<sub>2</sub>Se/KV<sub>2</sub>Se<sub>2</sub>O MTJ in two orientation alignments.** (a) Schematic atomic model of the unrotated device configuration; (b-c) Transmission spectrum and TMR values in (a); (d) Schematic model of device configuration where the K<sub>2</sub>Se barrier rotated 90° relative to that in (a); (e-f) Transmission spectrum and TMR values for rotated MTJ device.

**Tab. S3. The total free energy of MTJ devices.**

|                                     | Total free energy |
|-------------------------------------|-------------------|
| The device structure in Fig. S8 (a) | -25016.33811eV    |
| The device structure in Fig. S8 (d) | -24976.43354eV    |



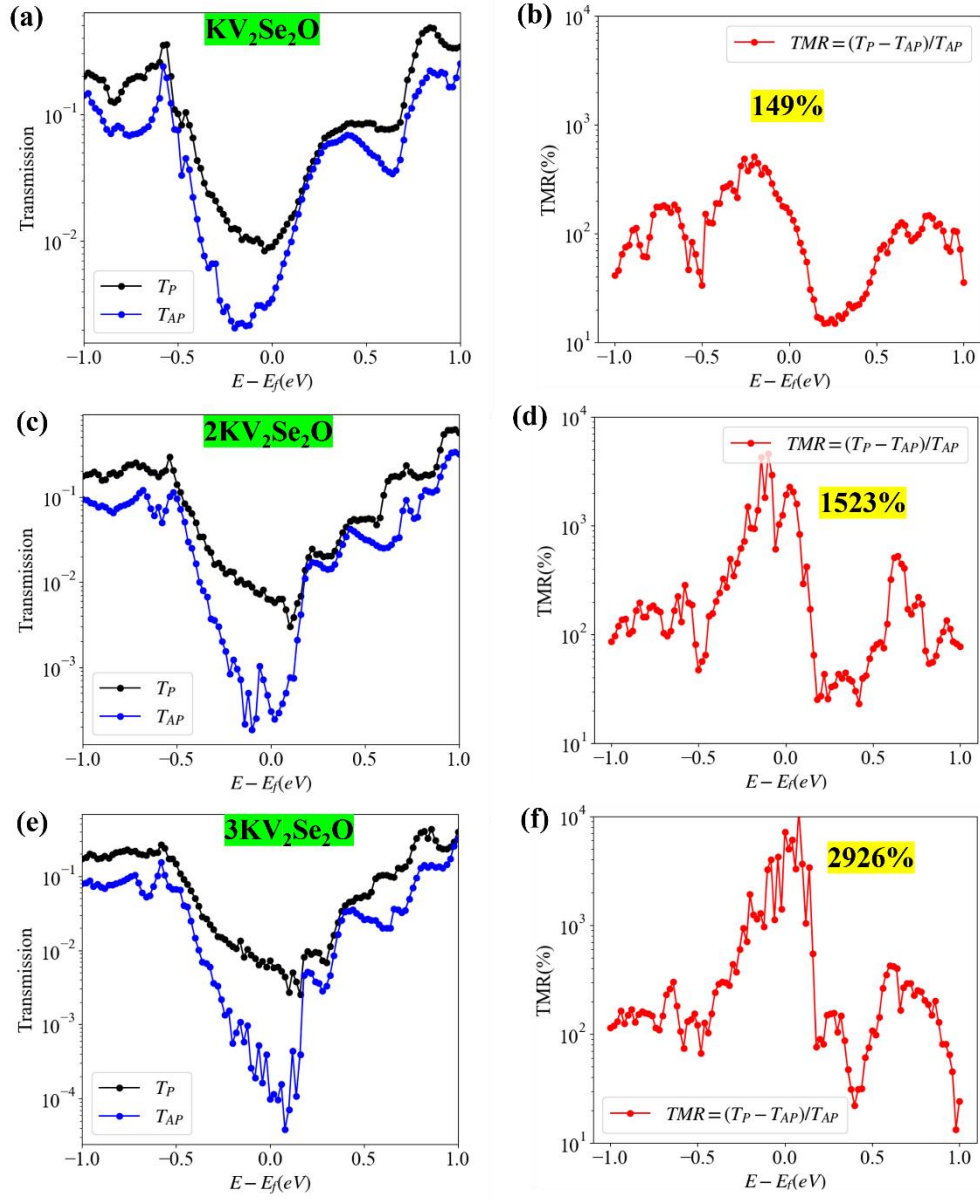
**Fig. S9.** The Projected Local Density of States for  $KV_2Se_2O$ -based MTJ devices in two distinct structural orientations. (a-b) The PLDOS for the two device structures in Fig. S8 (a, d).

**Tab. S4.** Transmission spectrum and TMR values of  $RbV_2Te_2O$ -based MTJ devices.

| RbV <sub>2</sub> Te <sub>2</sub> O-based |           | $T_P$    | $T_{AP}$ | TMR (%) |
|------------------------------------------|-----------|----------|----------|---------|
| Type-1                                   | Spin up   | 1.844e-5 | 1.252e-8 | 147160  |
|                                          | Spin down | 1.844e-5 | 1.252e-8 |         |
| Type-2                                   | Spin up   | 3.993e-5 | 6.347e-7 | 6190    |
|                                          | Spin down | 3.993e-5 | 6.347e-7 |         |

**Tab. S5.** The Transmission spectrum and TMR values of  $XV_2Y_2O/n$ -Layers  $X_2Y/XV_2Y_2O$  MTJ devices.

| Device type                                                                                                    | Layers | $T_P$    | $T_{AP}$  | TMR (%) |
|----------------------------------------------------------------------------------------------------------------|--------|----------|-----------|---------|
| KV <sub>2</sub> Se <sub>2</sub> O/ <i>n</i> -Layers<br>K <sub>2</sub> Se/ KV <sub>2</sub> Se <sub>2</sub> O    | 3      | 0.475e-4 | 5.015e-4  | 847     |
|                                                                                                                | 5      | 1.704e-4 | 1.157e-6  | 14633   |
|                                                                                                                | 7      | 1.131e-5 | 6.066e-7  | 1764    |
|                                                                                                                | 9      | 4.987e-7 | 3.059e-9  | 16199   |
|                                                                                                                | 11     | 9.738e-8 | 3.963e-10 | 24475   |
| RbV <sub>2</sub> Te <sub>2</sub> O/ <i>n</i> -Layers<br>Rb <sub>2</sub> Te/ RbV <sub>2</sub> Te <sub>2</sub> O | 3      | 9.450e-4 | 2.007e-5  | 4607    |
|                                                                                                                | 5      | 4.490e-5 | 1.957e-8  | 229311  |
|                                                                                                                | 7      | 1.959e-6 | 2.552e-8  | 7580    |
|                                                                                                                | 9      | 2.350e-7 | 6.153e-12 | 3819617 |
|                                                                                                                | 11     | 4.474e-8 | 2.536e-11 | 176292  |



**Fig. S10.** The transmission spectrum and TMR values in Pt/*n*-KV<sub>2</sub>Se<sub>2</sub>O/K<sub>2</sub>Se/*n*-KV<sub>2</sub>Se<sub>2</sub>O/Pt MTJ devices. (a,c,e) represent the transmission spectrum of MTJs with 1-layer, 2-layer, and 3-layer KV<sub>2</sub>Se<sub>2</sub>O; (b,d,f) represent the TMR values of MTJs with 1-layer, 2-layer, and 3-layer KV<sub>2</sub>Se<sub>2</sub>O. These results about KV<sub>2</sub>Se<sub>2</sub>O are calculated using QuantumATK with FHI pseudopotentials and a single-zeta-polarized basis set.

**Tab. S6. The transmission and TMR values of Ag/2- $XV_2Y_2O$ / $X_2Y$ /2- $XV_2Y_2O$ /Ag MTJ devices. The results are calculated using QuantumATK with PseudoDojo pseudopotentials, and SGGA+ $U$  ( $U=0.5$  eV).**

|              | $T_P$     | $T_{AP}$  | TMR (%) |
|--------------|-----------|-----------|---------|
| $X=K, Y=S$   | 9.248e-04 | 2.493e-07 | 370923  |
| $X=K, Y=Se$  | 5.982e-04 | 4.757e-07 | 125667  |
| $X=K, Y=Te$  | 2.316e-03 | 9.312e-05 | 2387    |
| $X=Rb, Y=S$  | 6.283e-04 | 9.361e-08 | 671051  |
| $X=Rb, Y=Se$ | 7.629e-04 | 7.513e-08 | 1015324 |
| $X=Rb, Y=Te$ | 1.684e-03 | 6.371e-05 | 2544    |
| $X=Cs, Y=S$  | 8.824e-04 | 4.380e-07 | 201353  |
| $X=Cs, Y=Se$ | 7.501e-04 | 1.029e-07 | 728662  |
| $X=Cs, Y=Te$ | 1.733e-03 | 4.951e-06 | 34895   |